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R. Gueroult and N.J. Fisch

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PRACTICALITY OF A PLASMA MASS FILTER FOR NUCLEAR FUEL REPROCESSING: SEPARATING LANTHANIDES FROM ACTINIDES *

R. Gueroult[ξ], N. J. Fisch
Princeton Plasma Physics Laboratory, PO Box 451, MS-29
Princeton, NJ 08543, USA

Abstract

There is a growing recognition [1,2] of the need for used nuclear fuel recycling technologies that are more proliferation-resistant alternatives to the Plutonium/Uranium refining by extraction (PUREX). However, the implementation of a closed fuel cycle, and more precisely of the minor actinides transmutation step, requires removing a priori the lanthanides. The chemical separation options remain limited because of the chemical similarity of americium with the lanthanides fission products. Separating lanthanides from actinides has therefore been labeled as one of the most difficult challenge in separation science [3].

Plasma filters offer an advantage over chemical solutions in that elements are dissociated. Each element can consequently be filtered without regard to chemical form. Thus, plasma mass filters have been recently proposed with the objective of nuclear waste remediation [4,5]. In particular, the capability of a new mass called the magnetic centrifugal mass filter has been studied in this context [6], highlighting the potential of plasma mass filters for nuclear waste remediation.

Here we analyze how such plasma filters could be helpful in separating lanthanides from actinides. More specifically, the influence of the elements mass shift as compared to the ones considered for nuclear waste remediation is investigated, with special care given to the modifications induced on the achievable plasma parameters. Estimations of achievable separation factor are obtained by means of numerical modeling.

I. RATIONALE FOR AN ACTINIDES / LANTHANIDES SEPARATION SCHEME

Reprocessing nuclear spent fuel has various motivations. Originally, the sole purpose was to extract fissionable Plutonium for producing nuclear weapons. With time, the focus of fuel reprocessing has moved to commercial reactors' spent fuel management cycle, enabling closed fuel cycle strategies, as opposed to the fuel direct disposal traditionally in effect in open fuel cycles. In this context, the main objective is the recovery of the unconsumed Uranium fuel (U-238), as well as Plutonium (Pu-239) produced by transmutation, for future use. The extracted Plutonium can then be recycled into Mixed OXyde (MOX) fuel, while the extracted Uranium could directly be re-used as fuel. This is currently achieved industrially by means of the PUREX [7] (Plutonium and Uranium Recovery by EXtraction) chemical process.

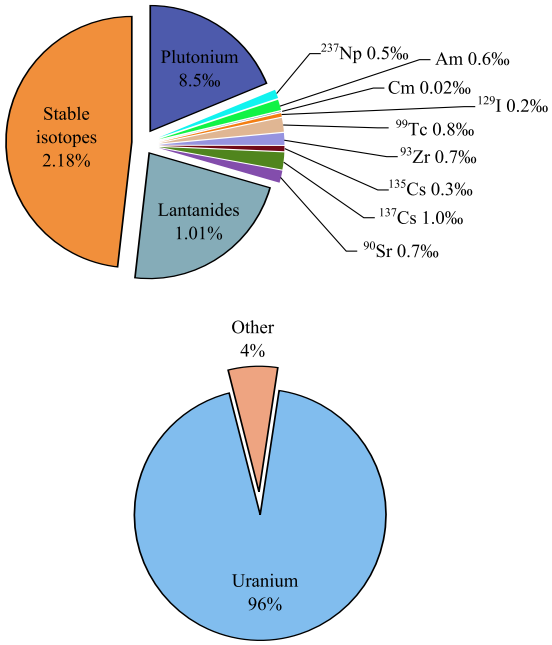

Figure 1. Typical composition of nuclear spent fuel.

Another motivation for nuclear spent fuel reprocessing is the reduction of the radioactivity levels, in particular through the reduction of high-level waste volume and mitigation of the long-term risks. The removal of unconsumed Uranium and Plutonium already offers in

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ξ email: rgueroul@pppl.gov

that sense a significant advantage since, as depicted in Fig. 1, it allows reducing the volume by up to 95%. The next step towards better waste management lies in the treatment of minor actinides and long lived fission products, which are on the long term responsible for most of the potential biological hazard, as indicated in Fig. 2.

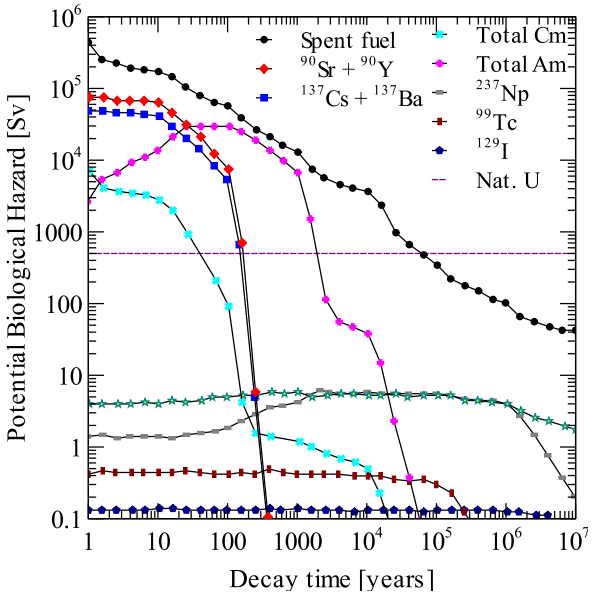

Figure 2. Breakdown of irradiated fuel potential biological hazard as a function of time for 1kg of irradiated actinides, from [8].

More specifically, it appears necessary to primarily remove Am, Cm, Cs and Sr. Because of the low neutron cross-section of ^{137}Cs and ^{90}Sr, no option but monitored storage exists for these elements after removal from other fission products. However, thanks to their limited half-lives (~30 years), the risks become limited after a few hundred years. On the other hand, the half-life of the main Americium isotope, Am-241, is about 432 years, and decays into Neptunium of which half-life is more than 2 million years. The solution considered is therefore transmutation. Because of the much larger neutron cross-section of Lanthanides, this step however requires removing a priori Am and Cm from Lanthanides. Because of the chemical proximity of Lanthanides to Actinides, a chemical separation is challenging [3]. This task is made even more complex by the relative amount of these species, namely about one Americium atom for twenty Lanthanides atoms as shown in Fig. 1. Once separated, Cm could be transmuted as well, but the preferred approach would consist in taking advantage of the relatively short half-life of the two mains isotopes (29 and 18 years for Cm-243 and Cm-244, respectively), and monitor their decay into Pu-239 and Pu-240. The naturally produced Pu isotopes could then be sent back into the fuel cycle.

In summary, transmutation of minor actinides found in nuclear spent fuel requires separating *a priori* these elements from the Lanthanides present in the waste stream. The chemical similarities existing between these elements make the removal of Am and Cm from the twenty times more abundant Lanthanides atoms hardly achievable through conventional chemical techniques. A non chemical alternative solution would consequently be desirable.

II. PLASMA MASS SEPARATION TECHNIQUES

Plasma separation in principle offers an advantage as compared to chemical techniques in that atoms are dissociated. Each atom can then be filtered without respect to its chemical form. In contrast, separation has to be obtained by means of differences among the intrinsic properties of the formed ions, such as the mass, the charge state, …, or any combination of these properties.

As an example, the idea of relying on plasmas to discriminate elements based on their mass has long been used, starting with the Calutron concept [9] during the Manhattan project in the 40's. These techniques however essentially remained geared towards isotopes separation. Only recently, plasma based separation has been considered for nuclear waste remediation [10]. Although the idea was to separate elements based on their mass, the separating principle consists in a threshold charge to mass ratio for ion confinement [4].

More recently, a new plasma mass filter concept has been proposed and analyzed in the same context [5,6]. In this concept, called the Magnetic Centrifugal Mass Filter (MCMF), separation is achieved by spinning a cylindrical collisional plasma with asymmetric confinement properties at each end. As shown in Fig. 3, confinement on the right side is achieved through a bending of the magnetic field lines toward the axis of rotation. Since the centrifugal force $m\Omega^2\mathbf{r}$ is proportional to the ion mass, for two ions of different masses and same parallel kinetic energy, the light ion will escape more favorably. The right side is consequently labeled the light element side. Confinement on the left side is achieved by means of a magnetic mirror. Magnetized ions following the magnetic field lines towards larger radii are reflected by the mirror force. However, heavy ions gain a larger parallel kinetic energy drifting to larger radial positions, while the mirror force $\mu\nabla B$, with $\mu = mV_\perp^{\,2}/(2B)$ the magnetic moment, is identical for two ions having the same perpendicular kinetic energy. Heavy elements will therefore exit more easily through the magnetic mirror, and the left side is consequently label the heavy element side. A direct consequence of these ion mass dependent confinement properties is the requirement for similar ion temperatures

for both ion species to be separated. The temperatures ratio has to be in any case smaller than the masses ratio for the mass selectiveness to occur [6]. Such a condition is however expected to be easily met thanks to inter species ion-ion collisions. Specifics about the working principles and practical operating conditions can be found in ref [5,6].

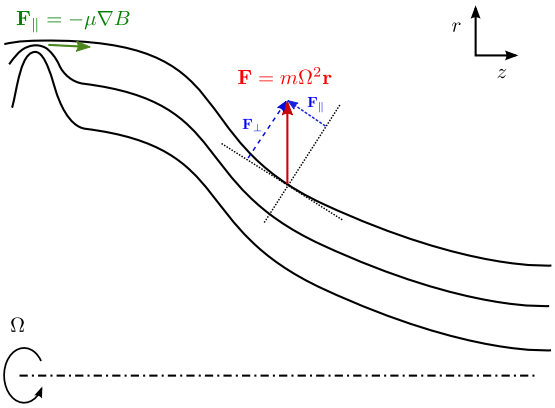

Figure 3. Separating principle in a Magnetic Centrifugal Mass Filter (MCMF): cut-view of the axi-symmetric configuration in the r-z plane. Full black lines denote the magnetic field lines. At one end, the confinement is dominated by magnetic forces, which are independent of mass, like in a magnetic mirror. At the other end, the confinement is dominated by centrifugal forces, which are proportional to the mass

Since the separation factor of rotating plasma configurations (plasma centrifuge, Archimedes Filter, MCMF) scales exponentially with the ions mass difference [11], such designs are poorly suited for isotopes separation. On the other hand, if looking at larger mass differences, it has been shown that such devices can in principle offer interesting throughputs [6, 10, 11]. We therefore consider here the potential of such filters, and particularly the MCMF, for the Lanthanides/Actinides separation challenge introduced in Sec. I.

III. AMERICIUM (Am-241) PLASMA BASED REMOVAL FROM LANTHANIDES

The separation challenge to be addressed, as discussed in Sec. I, consists in separating Actinides, and more particularly Am-241, from a significantly larger (up to 50 times) volume of Lanthanides, as pre-extracted from spent nuclear fuel fission products. A better picture of the mass distribution of Lanthanides within the waste stream can be obtained from the literature [12]. Such a distribution is plotted in Fig. 4. We see that even if the mass of elements within the Lanthanides series can in principle be as high as 176 amu with Lu-176, smaller masses are found within the fission products, with an average mass for the Lanthanides of about 144 amu.

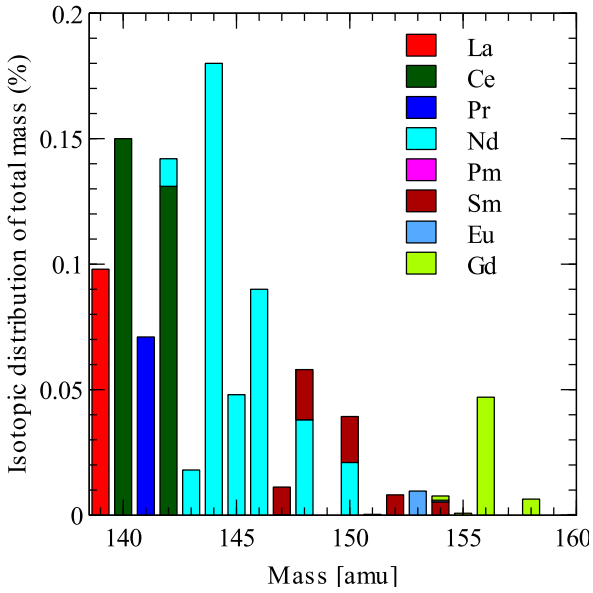

Figure 4. Isotopic distribution of lanthanides mass in nuclear spent fuel fission products, from [12]. The mean mass is about 144 amu.

Based on this Lanthanides mass distribution, we numerically study the separation of Am-241 from a test element of 144 amu. The full description of the numerical model used can be found in ref [6], and the magnetic field topology and strength are kept identical. Typical results obtained for a rotating velocity of about 4 km.s^{-1} (angular frequency of about 10^5 rad.s^{-1}) at the initial ion position are shown in Fig. 5. Note that because of the large mass of the considered ions, such a rotation speed is already larger than the critical ionization velocity (CIV) [13],

$$v_c = \sqrt{\frac{2e\varepsilon_i}{m}} \quad , \qquad (1)$$

with e the elementary charge, m the ion mass and ε_i the ionization energy in eV, as indicated in Tab. 1. Potential solutions to address this limitation, such as wave induced rotation, have however been proposed [5,6].

For the lower densities, the mass separation is clearly outlined. Quantitatively, for $n = 6.10^{11}$ cm^{-3}, the heavy to light side stream ratio is 1.33 for the test ion of 144 amu, while it is 3.15 for Am-241. Looking at the higher density case $n = 2.10^{12}$ cm^{-3}, these ratios become 0.86 and 1.41 for these two same ions. In addition, as the density increases, an increase of the radial losses is observed, with up to one out of ten ions being lost through this channel. This increase results from an increase of the collisionality,

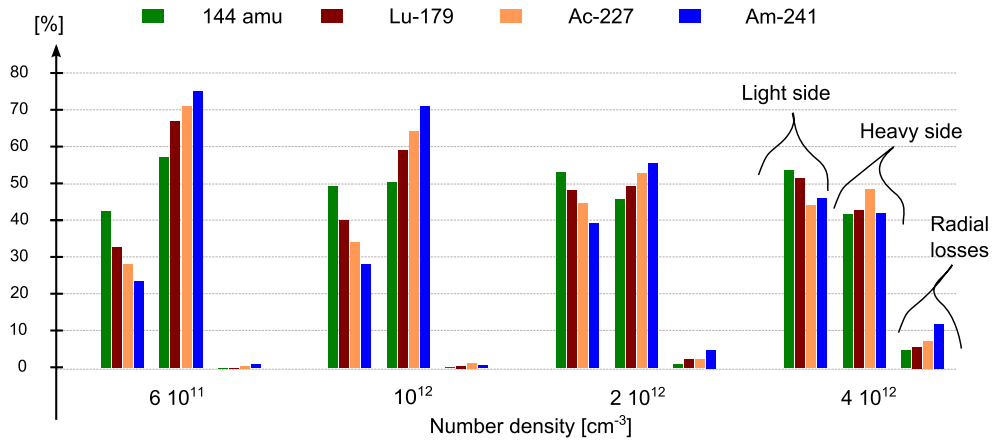

Figure 5. Ratios of particles collected on each end of the device, as well as lost radially, for various plasma number densities.

which increases the ion transport perpendicularly to the magnetic field lines, that is to say mainly radially. The fact that, for a given number density, the radial losses are larger for heavier elements is consistent with this trend. The radial losses levels remain however acceptable for number densities up to a few 10^{12} cm^{-3}.

Although these preliminary results indicate a mass selectivity for the particles of interest, underlining in turn the potential of plasma mass filters for the separation task considered in this paper, various questions remain to be addressed. One concern lies in the small volumic fraction of the particular ion to be removed (Am-241) from the much larger bulk stream of Lanthanides. This consideration is expected to modify the properties of ion-ion pitch angle scattering, and therefore the probability for a given ion to be extracted at one or the other end of the device. Another and more general reserve resides in the limited separation factor obtained in one pass. As a matter of fact, and even if an optimization of the magnetic field topology is expected to yield larger separation factors, the separation levels required are significantly larger than the ones likely to be achieved in one pass. This last point calls for sequential processing. To this respect, the Magnetic Centrifugal Mass Filter offers a significant advantage as compared to the Archimedes Filter [10] in that the waste is extracted along the field lines, making in principle a sequential processing easier. Specific methodologies are however yet to be developed

IV. SUMMARY

In this paper, we highlighted how plasma based mass separation could be useful with the objective of separating minor Actinides, in particular Am-241, from Lanthanides within the purpose of nuclear spent fuel reprocessing.

Because of the much larger neutron cross section of Lanthanides elements, removal of Am-241 from Lanthanides is required before transmutation of the long lived Am-241 isotope into shorter lived radionuclides. However, because of similar chemical forms, the separation of these elements through chemical processing is challenging.

Table 1. Critical ionization velocity (CIV) for various ions of interest

Ion	Mass [amu]	Ionization Energy [eV]	CIV [km.s^{-1}]
La	139	5.58	2.77
Ce	140-142	5.54	2.74-2.72
Pr	141	5.47	2.71
Nd	144	5.53	2.70
Eu	153	5.67	2.65
Gd	156	5.60	2.61
Ac	227	5.18	2.09
Am	241	6.00	2.18

Plasma mass filters, which have recently been shown to offer interesting properties for the purpose of nuclear waste remediation, could present an appealing potential within this context too. More particularly, numerical simulations indicate that the Magnetic Centrifugal Mass Filter could be suitable for this task. Mass selectivity is numerically demonstrated for the ions of interest and realistic operating conditions.

Although these results suggest that plasma mass filtering could be useful to address challenging separation tasks as encountered in nuclear waste reprocessing, various considerations remain to be examined. Sequential processing capabilities appear as one of the directions to investigate primarily.

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Information Services
Princeton Plasma Physics Laboratory
P.O. Box 451
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